

# Giant-diamagnetic and magnetization-step effects in HgMnTe monocrystal

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## Abstract

In  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$  ( $x \geq 0.16$ ) monocrystal, the giant-diamagnetic (GDM) and magnetization-step phenomena have been observed in spin glass (SG) regime. The susceptibility of GDM is about 100-1000 times than that of classic diamagnetic. It can be interpreted that: due to the long-range antiferromagnetic (AF) exchange interactions and the non-uniform random distribution of  $\text{Mn}^{2+}$  ions in  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ , a quasi-static spin wave forms and produces the GDM phenomenon below the critical temperature and magnetic field. Meanwhile, this theory is proved by Monte Carlo simulations in a two-dimensional AF cluster based on XY model. Hence, it is possible to emerge long-range magnetic order structure in SG state.

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At low temperature, it is well known that the combined effects of randomness and frustration may lead to spin-glass (SG) behavior in disordered spins system, such as magnetic alloy, magnetic oxides and semimagnetic semiconductor (SMSC or DMS). [1, 2] For the SG of metallic alloy or magnetic metal oxides, it is difficult to separate the contribution of the conduction electrons from that of the localized spins, liking RKKY mechanism. Therefore, for a better understanding of SG, Mn-based SMSCs are appropriate candidates for studying SG in experiment, due to pure antiferromagnetic (AF) exchange interaction and very low carriers concentration. In SG state, the global ground state of system always is to a major concern problem and not be resolved until now. Generally, mostly classical SG theories based on mean-field theory and short-range AF exchange (e.g., Ising model and Sherrington-Kirkpatrick model[3]) suppose that the spins have no long-range magnetic order but instead have frozen or quasi-static orientations which vary randomly over macroscopic distances at low temperatures.[1] Meanwhile, it predicts the limit concentration of the SG transition is about 17% in SMSC with fcc structure.[4]

Whereas, in  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$  (fcc structure), the existing results of magnetic and specific heat experiments have proved that: (i) the AF exchange interaction between  $\text{Mn}^{2+}$  ions contains long-range exchange mechanisms, such as Bloembergen-Rowland exchange;[5–7] (ii)  $\text{Mn}^{2+}$  ions are not exactly random uniform distribution but random fluctuation distribution in space.[8–10] Both features go against the basic hypothesis of classical SG theories. Can it produce some new effects on the spin arrangement of SG state, liking long-range magnetic ordered structures? From a fundamental perspective, this is a very important issue in SG theory. For this purpose, we investigate the magnetic properties of  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$  with variant Mn concentrations, particularly near the SG regime.

In this work, the DC field susceptibility (2–300 K) and magnetization (2–10 K) measurements with physical property measurement system (PPMS) of Quantum Design were carried out on four  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$  monocrystal samples grown by modified Bridgman method and annealed in Hg vapor. Four samples denoted as NO1 ( $x \simeq 0.1$ ), NO2 ( $x \simeq 0.16$ ), NO3 ( $x \simeq 0.191$ ) and NO4 ( $x \simeq 0.207$ ) respectively, where NO4 was the best monocrystal. Zero field cooled (ZFC) method was adopted in the susceptibility measurements and the time interval for each measuring point was 1 sec. In addition, Hall measurements proved four samples were strong p-type and hole concentrations were lower than  $10^{14}\text{cm}^{-3}$  below 10 K.

Figure 1 presents the magnetization curves of four samples at different temperatures.

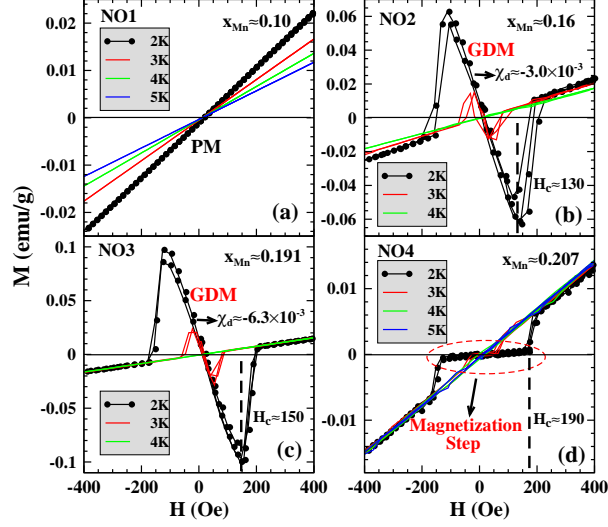


FIG. 1: The temperature-dependent magnetization curves of  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$  monocrystals with different Mn concentrations. These results show that when Mn concentration approaches or exceeds the limit of SG transition (17%) in fcc structure, GDM and magnetization-step emerge below the critical temperature ( $T_c$ ) and magnetic field ( $H_c$ ). The susceptibility of GDM is about 100–1000 times than that of classic diamagnetic. In addition, the values of  $T_c$  and  $H_c$  go up as Mn concentration increases.

From 2 K to 5 K, the magnetization curves of NO1 are simple straight lines indicating good paramagnetic (PM) state, as shown in Figure 1 (a). However, the remaining samples (NO2, NO3 and NO4) emerge novel and interesting magnetization phenomena at low temperatures, as illustrated in Figure 1(b), (c) and (d). For NO2 and NO3, both of magnetization curves show giant-diamagnetic (GDM) phenomenon below 4 K (called the critical temperature  $T_c$ ). The following are the master features of GDM: (1) The absolute value of GDM susceptibility is very large and depends on Mn concentration ( $\chi_d = -3.0 \times 10^{-3}$  for NO2 and  $\chi_d = -6.3 \times 10^{-3}$  for NO3), which is about 100–1000 times greater than that of classic diamagnetic ( $\chi_d = -10^{-6} \sim -10^{-5}$ ). (2) There is a critical magnetic field ( $H_c$ ) for the existence of GDM at fixed temperature, e.g.,  $H_c \simeq 130$  Oe for NO2 and  $H_c \simeq 150$  Oe for NO3 at 2 K. As magnetic field exceeds  $H_c$ , GDM state rapidly changes into paramagnetic state. (3)  $H_c$  decreases with temperature rising. As regards NO4, when temperature lower than 5 K (about the  $T_c$ ), its magnetization curves emerge magnetization-step instead of GDM. Meanwhile, this step also only exists under a critical magnetic field (e.g.,  $H_c \simeq 190$  Oe at 2 K), and gradually disappears as temperature rises.

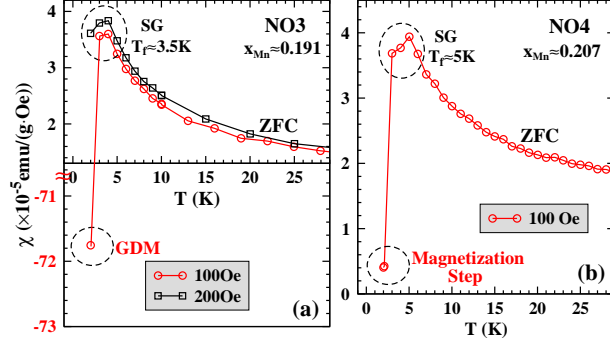


FIG. 2: The susceptibility curves ( $\chi$ - $T$ ) of NO3 and NO4 measured by ZFC method. (a) shows the  $\chi$ - $T$  curves of NO3 at 100 Oe and 200 Oe. Both curves emerge a cusp structure at 3.5 K, which means SG transition. Meanwhile, at 100 Oe, the curve appears large negative value (GDM) below 3 K. (b) is the  $\chi$ - $T$  curve of NO4 at 100 Oe which emerges both SG transition (5 K) and magnetization-step (below 3 K).

Comparing the results of magnetization measurements in four  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$  samples, it is clear that: (i) When Mn concentration approaches or exceeds the limit of spin-glass transition (17%) in fcc structure, GDM and magnetization -step will appear below the critical temperature ( $T_c$ ) and magnetic field ( $H_c$ ); (ii) As Mn concentration increases, the  $T_c$  and  $H_c$  of GDM and magnetization -step also slowly go up.

In order to making a further justification for GDM and magnetization-step, the susceptibility measurements with the ZFC method were carried out on NO3 and NO4. Figure 2 shows the susceptibility curves ( $\chi$ - $T$ ) of NO3 and NO4 from 2 K to 30 K at weak magnetic fields ( $H=100$  Oe and 200Oe). For NO3, its  $\chi$ - $T$  curves emerge a cusp structure at about 3.5 K (exactly  $T_c$ ), which means the occurrence of SG transition, as shown in Figure 2(a). In the SG regime, the value of susceptibility markedly changes with magnetic field, which are positive value at  $H=200$  Oe (greater than  $H_c$  at 2 K), but appears large negative value (corresponding to GDM) at  $H=100$  Oe (less than  $H_c$  at 2 K). As well, the  $\chi$ - $T$  curve of NO4 at 200 Oe magnetic field also emerges SG transition at 5 K ( $T_c$ ) and the rapidly reduction of susceptibility below 5 K (corresponding to magnetization-step), as illustrated in Figure 2(b). Hence, the results of susceptibility curves prove the existence of GDM and magnetization-step again. More importantly, the GDM and magnetization-step are associated with the SG transition.

Generally, the classical diamagnetic comes from electron orbit precession and electro-

magnetic induction, the value of which is independent of temperature and magnetic field. But the GDM of  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$  is related to temperature and magnetic field. In addition, the relaxation magnetization model of SG proposed by Lundgren, which assumes a uniform random distribution and short-range AF interactions of magnetic ions, only leads to positive susceptibility (paramagnetic) in DC magnetic measurements.[11, 12] So, what are the reasonable physical mechanisms of GDM and magnetization-step in  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ ?

We think these novel magnetization phenomena should come from the effects of strong long-range AF interactions between  $\text{Mn}^{2+}$  ions in  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ . The following are our reasons in details. In the magnetization process, whether a spin of  $\text{Mn}^{2+}$  can be flipped by magnetic field depends on the competition of three factors: thermal fluctuation, magnetic field and AF interactions between  $\text{Mn}^{2+}$  ions. Usually, thermal fluctuation leads to spins chaotic flipping, magnetic field causes the orderly arrangement of spins and AF interactions make spins frozen. The condition of whether a spin is free or magnetic frozen is[13]

$$\sum_j J_{nn} S_{\text{Mn}}^2 \hat{s}_i \cdot \hat{s}_j \geq \frac{3}{2} k_B T + g_{\text{Mn}} \mu_B S_{\text{Mn}} H \quad (1)$$

where  $\sum_j J_{nn} S_{\text{Mn}}^2 \hat{s}_i \cdot \hat{s}_j$  is the AF exchange energy with the nearest neighbors,  $\frac{3}{2} k_B T$  is the thermal kinetic energy and  $g_{\text{Mn}} \mu_B S_{\text{Mn}} H$  is the magnetization energy.

At high temperatures, due to strong thermal fluctuations, Eq.1 can not be satisfied even at zero magnetic field. Thus, the spins of  $\text{Mn}^{2+}$  ions freely rotate and can be overturned easily by magnetic field. However, at low temperatures, thermal fluctuations become weak and Eq.1 is easy to satisfied, especially for high Mn concentration and strong long-range AF interactions between  $\text{Mn}^{2+}$  ions. In this case, the spins of  $\text{Mn}^{2+}$  ions are mostly frozen and form AF clusters, as shown in Figure 3(a).

Then, according to Eq.1, when Mn concentration with uniform random distribution approaches or exceeds a critical value, it needs the same nonzero magnetic field for all  $\text{Mn}^{2+}$  ions to break down Eq.1 and generate magnetization below a critical temperature ( $T_c$ ). In other words, magnetization can not appear when magnetic field and temperature are both less than the critical values ( $H_c$  and  $T_c$ ) in high Mn concentration area, as shown in NO4 sample. This is the physical mechanism of magnetization-step in  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ .

As to the physical mechanism of GDM, it involves two key factors. One stems from the effect of non-uniform random distribution of  $\text{Mn}^{2+}$  ions in  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ . When the distribution of  $\text{Mn}^{2+}$  is inhomogeneous in space, the magnetized conditions (Eq.1) of  $\text{Mn}^{2+}$

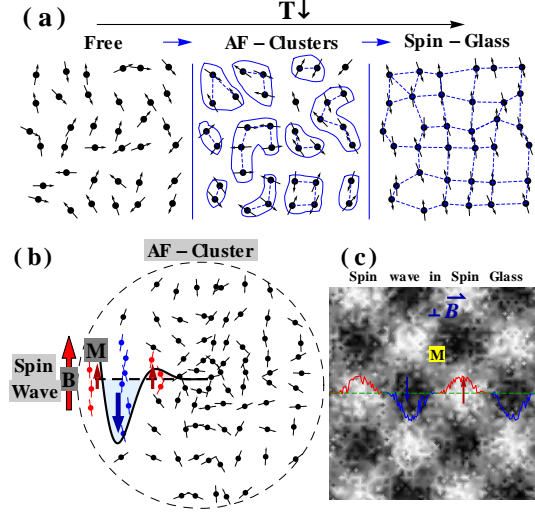


FIG. 3: (a) shows the formation of AF clusters and spin-glass as temperature decreases. (b) shows the structure of quasi-static spin wave in a 2D AF cluster with long-range AF interactions and non-uniform random distribution of  $Mn^{2+}$  ions. (c) shows the possible interconnect structure of quasi-static spin waves in SG state.

ions will be different from each other. It leads to, at the same temperature and magnetic field, the spins in high concentration region are easy to frozen, but in low concentration region are easily magnetized. As a result, the spin arrangement of  $Mn^{2+}$  ions is also inhomogeneous in space, especially in AF cluster which can be taken as the unit to compute the spin arrangement of ground state in non-uniform random distributing spin system.

The other is due to the strong long-range AF interactions between  $Mn^{2+}$  ions in  $Hg_{1-x}Mn_xTe$ . In high Mn concentration area, this will cause that the AF exchange energy with further neighbors ( $J_{fn}(R)S_{Mn}^2$ ) is stronger than thermal kinetic energy of  $Mn^{2+}$  ion when temperature below a critical value. Consequently, the spins of  $Mn^{2+}$  ions emerge multi-frustration effect, which exist not only between the nearest neighbors but also with further neighbors. strongly correlated

Combining the effects of two factors, a spin flip of  $Mn^{2+}$  ion occurred at the edge of AF cluster can impact the spin arrangement of both the nearest neighbors and further neighbors, and then produce a series of chain reactions in AF cluster, liking "dominoes" effect. At the right temperature and magnetic field, this effect can create a quasi-static spin wave with oscillating spin arrangement, as presented in Figure 3(b). More importantly, the amplitude of spin waves also depends on the Mn concentration, which may lead to a net negative

magnetic moment (opposite to magnetic field direction) in non-uniform random distributing AF clusters. Meanwhile, in the SG state, AF clusters are interconnect with each other forming a AF "super-cluster". Hence, the quasi-static spin waves can propagate in space, causing the GDM phenomenon in  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ , as shown in Figure 3(c). This quasi-static spin wave caused by local spins is similar to the spin-density wave (SDW) caused by electrons in chromium alloys.[14]

It is hard to strictly confirmed the model of quasi-static spin waves by analytical method. However, it is possible to verify the rationality of this model by numerical simulation. For this purpose, Monte Carlo (MC) simulation was employed to compute the spin configuration of a two dimensional (2D) AF cluster based on XY model at different temperatures and magnetic fields. Moreover, in order to overcoming the influence of metastable as much as possible, simulated annealing was applied to seek the ground state of 2D AF cluster.[15]

As an example, the simulation results of a 2D AF cluster referring to the properties of  $\text{Hg}_{0.809}\text{Mn}_{0.191}\text{Te}$  (NO3 sample) are discussed in this paper.[16, 17] This 2D AF cluster is taken as circular shape (the diameter is about 32.52 nm) and contains 399  $\text{Mn}^{2+}$  ions with non-uniform random distribution along the radial direction. In the central region, the average distance between the nearest  $\text{Mn}^{2+}$  ions ( $\bar{a}_{nn}$ ) is about 1.163 nm. From the center to the edge,  $\bar{a}_{nn}$  decreases linearly, which is about 1.221 nm in the edge region. In  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ , the exchange function of long-range AF interactions between  $\text{Mn}^{2+}$  ions is taken as  $J(R_{ij}) = J_1(\bar{a}_{nn}/R_{ij})^5$ , where  $R_{ij}$  is the distance between two  $\text{Mn}^{2+}$  ions and  $J_1(= -7.0\text{K}_B)$  is AF exchange integral constant between the nearest neighbors  $\text{Mn}^{2+}$  ions.[13, 18] The Hamiltonian of each  $\text{Mn}^{2+}$  ion is in form of Eq. 2. In MC simulations, the truncation radius  $R_{cut}=5\bar{a}_{nn}$ ,  $S_{Mn}=5/2$ ,  $g_{Mn}=2$ , and the number of MC steps is  $10^5$  at each temperature point.

$$H_i = - \sum_{R_{ij} \leq R_{cut}} J(R_{ij}) S_{Mn}^2 \hat{s}_i \cdot \hat{s}_j - g_{Mn} \mu_B \vec{B} \cdot \vec{S}_i \quad (2)$$

Figure 4 show the typical MC simulation results of the 2D AF cluster, including the  $\bar{M}_y$ -T curves ( $\bar{M}_y$  denotes the average magnetic moment of all  $\text{Mn}^{2+}$  ions along magnetic field direction) and the spin configurations of ground state at different temperatures and magnetic fields. When magnetic field is 100 Gs, the  $\bar{M}_y$ -T curve emerges both cusp structure (SG transition) and negative value (paramagnetic-diamagnetic transition below 4 K), moreover, the spin configurations of ground state prove that a quasi-static spin wave really exists and

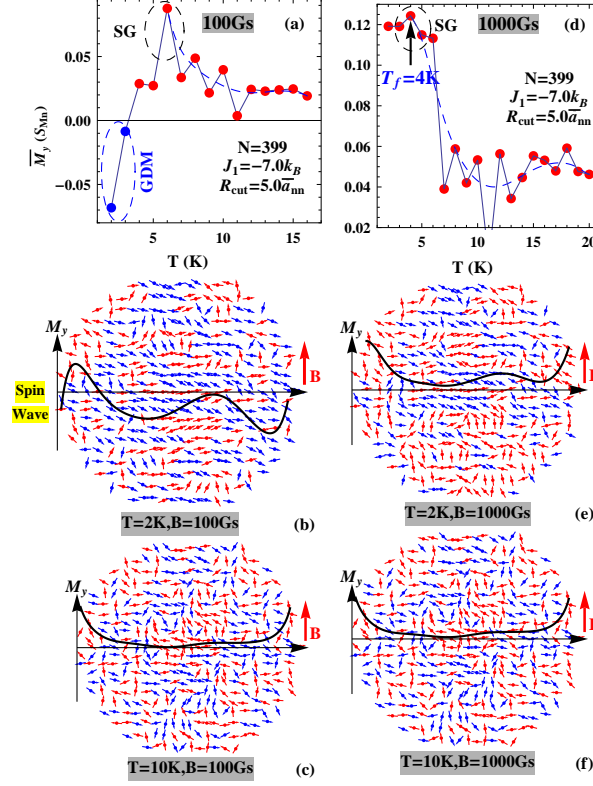


FIG. 4: The typical MC simulation results of a 2D AF cluster with long-range AF interactions and non-uniform random distribution. At 100 Gs, the  $\bar{M}_y$ - $T$  curve emerges SG transition (about 5 K) and GDM (below 4 K) for (a). Meanwhile, the spin configurations of ground state show a quasi-static spin wave which changes with temperature for (b) and (c). However, at 1000 Gs, the  $\bar{M}_y$ - $T$  curve only emerges SG transition and the quasi-static spin wave is faint in the 2D AF cluster, for (d), (e) and (f).

gradually disappears as temperature rises, as shown in Figure 4(a), (b) and (c). However, when magnetic field increases to 1000 Gs, the  $\bar{M}_y$ - $T$  curve only emerges cusp structure, and the quasi-static spin wave is faint or absent which is insufficient to cause paramagnetic-diamagnetic transition at low temperatures, as shown in Figure 4(d), (e) and (f).

These MC simulation results of the 2D AF cluster demonstrate that: a quasi-static spin wave really exists and leads to paramagnetic-diamagnetic transition (similar to the GDM in experiments) below the critical temperature, then gradually vanishes as temperature or magnetic field increases. Meanwhile, these results are basically consistent with the experimental results of GDM in  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$  (NO2 and NO3 samples). Hence, in  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ , the model of quasi-static spin waves inducing GDM is reasonable.

In conclusion, the results of DC magnetic measurements prove  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$  ( $x \geq 0.16$ ) monocrystals emerge giant-diamagnetic (GDM) and magnetization-step when temperature and magnetic field below the critical values ( $T_c$  and  $H_c$ ). The susceptibility of GDM is about 100-1000 times than that of classic diamagnetic. These novel magnetic phenomena come from the effects of long-range AF exchange interactions and non-uniform random distribution of  $\text{Mn}^{2+}$  ions in  $\text{Hg}_{1-x}\text{Mn}_x\text{Te}$ , such as inducing a quasi-static spin wave which produces the GDM. In addition, in a 2D AF cluster with long-range AF interactions and non-uniform random distribution, Monte Carlo simulations confirm that quasi-static spin waves really exist, which lead to paramagnetic-diamagnetic transition in the SG regime and gradually disappear as temperature or magnetic field increases. Hence, it is possible to emerge long-range magnetic order structure in SG state.

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